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## Identification of Sources of Phoenix Aerosol by Positive Matrix Factorization

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### Abstract

Chemical composition data for fine and coarse particles collected in Phoenix, AZ were analyzed using positive matrix factorization (PMF). The objective was to identify the possible particle sources contributing mass at the sampling site. PMF uses estimates of the error in the data to provide optimum data point scaling and permits a better treatment of missing and below-detection-limit values. It also applies the non-negativity constraints to the factors. Two fine particle data sets were available because samples were collected with two different samplers; a conventional dichotomous sampler and a DRI dual fine particle sequential sampler (DFPSS). Six source factors were obtained from both fine particle data sets and have been identified as possible aerosol sources for the fine particles. They are: (1) motor vehicles described by the high concentrations of organic carbon (OC) and elemental carbon (EC) with some soil dust components, (2) biomass burning which is typified by high concentrations of OC, EC and K, (3) copper smelter characterized by S, Cu, Zn, and Pb, (4) sea salt factor dominated by Na and Cl, (5) soil factor represented by Al, Si, Ca, Ti and Fe, and (6) S-factor (secondary sulfate) which is likely to represent coal power plant emissions. For the coarse particles collected in the dichotomous sampler, a 5-factor model gave the most satisfactory source profiles. The identified possible sources are: (1) sea salt, (2) soil dust, (3) Fe source/motor vehicle, (4) construction (high Ca), and (5) coal power plant. To estimate the mass contributions of the resolved sources, the particulate matter mass was regressed against the factor scores was performed. It was found that the major sources for the fine particles were the motor vehicle and the biomass burning factors. These two sources contributed had higher concentrations in winter time. For the coarse particles, the major source contributions were soil and construction (high Ca). These sources also peaked in winter time.

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## INTRODUCTION

It is of great importance to identify air pollution source characteristics in the development of air quality control strategies. Receptor modeling, using measurements of aerosol chemical compositions at a sampling site, is often a reliable way to provide information regarding source characteristics [1].

Multivariate receptor models are based on the analysis of the correlation between measured concentrations of chemical species assuming that highly correlated compounds come from the same source. One commonly used multivariate receptor model is principal component analysis (PCA) [2]. PCA extracts the principal components explaining the majority of variance of the data matrix that are then qualitatively interpreted as possible sources. Although PCA has been applied as a tool for source identification in some air quality studies [3,4], it suffers from several drawbacks in general. The factors of PCA are rarely physically explainable without further transformation (rotation), and no fully satisfactory rotation techniques have yet been found. In addition, PCA really represents a least squares (LS) minimization. This fit is weighted by implicitly assuming unrealistic standard deviations for the variables in the data matrix. Thus, the result of a principal component analysis does not represent a minimum variance solution since it is based on incorrect weighting [5,6]. Among other limitations, PCA can not properly handle missing and below-detection-limit data, a common occurrence in environmental measurements.

A new approach called Positive Matrix Factorization (PMF) has been developed by Paatero [7,8] to address the abovementioned problems. In PMF, error estimates for each data value are utilized as point-by-point weights. Such weighting scheme allows the inclusion of uncertain data in the analysis by giving them low weights. In addition, constraints on the results such as non-negativity of the factors are integrated into the computational process. These features make physical sense in receptor modeling of environmental data. PMF has been successfully applied to the analysis of PTEAM (Particle Total Exposure Assessment Methodology) data from Riverside, California [9], for the source identification of bulk wet deposition in Finland [10], of aerosol in Alaska [11], and of Arctic aerosol [12]. Paterson *et al.* [13] applied PMF to air quality and temperature data collected at a series of sites around the southern end of Lake Michigan in 1997 and used three factors to reproduce 75% of the variation in the data. Lee *et al.* [14] have applied PMF to urban aerosol compositions in Hong Kong. They were able to identify up to 9 sources that provided a good apportionment of the airborne particulate matter.

To examine the applicability of this new type of factor analysis as a source identification tool, PMF was performed on the Phoenix aerosol data. This paper presents the results of PMF study of Phoenix aerosol chemical composition data for fine and coarse particles. The major objective was to

identify possible aerosol sources at the sampling site, Phoenix using two different samples. Through the regression of the particulate matter mass against the factor scores, the mass contributions of the resolved sources were estimated and compared, which provided better understanding of the origin and impact of Phoenix aerosol.

## **DATA DESCRIPTION**

The location of sampling site is Phoenix, AZ (3847 West Earl Drive, Latitude: N33°48'46", Longitude: W112°14'17", Elevation: 1007 ft). There were two different airborne particle samplers. The dual fine particle sequential sampler (DFPSS) developed at the Desert Research Institute and described in the next subsection. The other sampler was a conventional dichotomous sampler that uses virtual impaction to separate the PM<sub>2.5</sub> from the coarse particulate matter.

### **DFPSS Data Set**

Daily, integrated 24-hour samples were collected on 37 millimeter (mm) diameter Teflon and quartz filter media for fine particle mass and species measurements using a dual fine particle sequential sampler (DFPSS). The DFPSS has two separate channels, each with separate, Teflon coated 2.5 : m cyclone inlets through which samples are collected simultaneously at a flow rate of 16.7 lpm. The inlets were located outside the monitoring shelter. The filter packs and the Teflon-coated distribution manifolds were located inside the shelter and maintained at 30 /C. The PM<sub>2.5</sub> cyclone on the DFPSS was replaced with a WINS impactor on 12/20/96. The shape of cut-point is sharper for the WINS compared to the cyclone inlet. The DFPSS is capable of operating unattended for four daily 24-hour sample pairs. Samples were collected every day, starting at 7:00 a.m. and ending at 7:00 a.m. the following day. The DFPSS collected daily samples on Teflon and quartz filter media (Teflon for mass and XRF analysis and quartz for volatilizable or organic carbon (OC), and elemental carbon (EC)).

Two energy dispersive X-ray spectrometers were used to produce the chemical elemental concentration data, a custom-made machine from Lawrence Berkeley Laboratories (LBL) and a commercially available Kevex (KEV) system. Both XRF instruments employed multiple choices for secondary excitation and utilized a helium atmosphere rather than vacuum in order to preserve volatile species. The quartz filters collected with the DFPSS were analyzed by Sunset Laboratory, Forest Grove, OR using the thermal optical transmission technique. This technique measured both organic carbon (OC)

and elemental carbon (EC). The samples were collected during the time period from March 1995 through June 1998. A total of 981 samples were finally obtained. Each sample was characterized by the measured concentrations of the following 46 chemical elements: Na, Mg, Al, Si, P, S, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Rb, Sr, Y, Zr, Mo, Rh, Pd, Ag, Cd, Sn, Sb, Te, I, Cs, Ba, La, W, Au, Hg, Pb, OC and EC. This 981 x 46 data matrix was used as the basis for the PMF study to infer possible aerosol sources. In this data matrix, there were some missing values (e.g. the LBL instrument could not quantify the elements Na and Mg due to the instrument design) and below-detection-limit values. The analytical uncertainty estimates associated with each measured concentration and the detection limits for both instruments were reported.

### DICHOT Data Set

Every third day collection of 24-hour integrated fine and coarse particle samples were made using a conventional dichotomous sampler (Andersen Instruments, Inc., Smyrna, GA). Integrated 24-hour PM<sub>2.5</sub> and PMCF (1.5 : m < d < 10 : m) particle samples were collected on Teflon filters. Na, Mg, Organic carbon (OC) and elemental carbon (EC) were not measured. In order to compare the PMF results for the two different sampler data sets for the fine particles, the measured concentrations of Na, OC and EC from the DFPSS study were used based on the corresponding dates. The Dichot samples were collected between June 1996 and June 1998. A total of 217 samples were obtained.

### DATA ANALYSIS

Positive Matrix Factorization (PMF) was used for the data analysis. PMF is a relatively new approach to solving the multivariate receptor modeling problem with a least squares approach [5]. Suppose  $\mathbf{X}$  is a  $n$  by  $m$  data matrix consisting of the measurements of  $n$  chemical species in  $m$  samples. The objective of multivariate receptor modeling is to determine the number of aerosol sources,  $p$ , the chemical composition profile of each source and the amount that each of the  $p$  sources contributes to each sample. The factor analysis model can be written as:

$$\mathbf{X} = \mathbf{GF} + \mathbf{E} \quad (1)$$

where  $\mathbf{G}$  is a  $n$  by  $p$  matrix of source chemical compositions (source profiles) and  $\mathbf{F}$  is a  $p$  by  $m$  matrix of source contributions (also called factor scores) to the samples. Each sample is an observation along the time axis, so  $\mathbf{F}$  describes the temporal variation of the sources.  $\mathbf{E}$  represents the part of the data variance un-modeled by the  $p$ -factor model.

In PMF, sources are constrained to have non-negative species concentration, and no sample can have a negative source contribution. The error estimates for each observed data point were used as point-by-point weights. The essence of PMF can thus be presented as:

$$\min_{\mathbf{G}, \mathbf{F}} Q(\mathbf{X}, \sigma, \mathbf{G}, \mathbf{F}) \quad (2)$$

where

$$Q = \left\| \frac{(\mathbf{X} - \mathbf{GF})}{\sigma} \right\|_{\mathbf{F}, \mathbf{G}}^2 = \sum_i \sum_j \left( \frac{e_{ij}}{\sigma_{ij}} \right)^2 \quad (3)$$

$$e_{ij} = x_{ij} - \sum_{k=1}^p g_{ik} f_{kj} \quad (4)$$

with  $g_{ik} \geq 0$  and  $f_{kj} \geq 0$  for  $k = 1, \dots, p$ , and  $\mathbf{F}$  is the known matrix of error estimates of  $\mathbf{X}$ . Thus, this is a least squares problem with the values of  $\mathbf{G}$  and  $\mathbf{F}$  to be determined.

This form of factorization is quite different from PCA.  $\mathbf{G}$  and  $\mathbf{F}$  are determined so that the Frobenius norm of  $\mathbf{E}$  divided by  $\mathbf{F}$  (point-wise) is minimized. As shown by Paatero and Tapper [8], it is impossible to perform factorization using a singular value decomposition (SVD) on such a point-by-point weighted matrix. PMF uses a unique algorithm in which both  $\mathbf{G}$  and  $\mathbf{F}$  matrices are varied simultaneously in each least squares step. The algorithm was described by Paatero [5].

Application of PMF requires that error estimates for the data be chosen judiciously so that the estimates reflect the quality and reliability of each of the data points. This feature provides one of the most important advantages of PMF, the ability to handle missing and below-detection-limit data by adjusting the corresponding error estimates. In the present application, over 70% of Na and Mg data points were missing due to the LBL instrument design, and there were some below-detection-limit data for other chemical elements indicated as negative values because of blank correction. The error estimate for each measured concentration was constructed using the combination of the analytical measurement uncertainty and 20% of detection limit value. Such a combination scheme, suggested in a previous study [11], appeared to work well in the present application. For each element, the missing data and below-detection-limit data were replaced by the geometric mean of the measured concentrations and half of the detection limit, respectively, and large error estimates were used for such values (4\*geometric mean and

detection limit, respectively). To reduce the effect of extreme values or outliers on the model performance, the robust PMF mode has been used.

## RESULTS AND DISCUSSION

### DFPSS Data

A critical step in PMF analysis is the determination of the number of factors. Analysis of the goodness of model fit,  $Q$ , as defined in Equation (3), can be used to help determine the optimal number of factors. Assuming that reasonable error estimates of individual data point are available, then fitting each value should add one to the sum and the theoretical value of  $Q$  should be equal to the number of data points in the data set. However, the resulting solutions also have to make physical sense within the system being studied. In real environmental data cases, there are often missing and below-detection-limit data points, their error estimates are mainly based on the investigator judgements, thus it is reasonable for the calculated  $Q$  value to deviate from the theoretical value to some extent. Also, it is always good practice to experiment with different numbers of factors and compare the analysis results. The results of the multiple linear regression calculation of the reconstructed particulate matter mass concentrations can provide additional measure of the quality of the fitted model by ensuring that the regression coefficients are positive.

The imposition of non-negativity constraints on the factors decreases the rotational freedom and, in some cases, produces unique solutions with no rotational freedom. However, some rotational ambiguity generally remains in the PMF solutions. In this application, rotational freedom existed. The acceptable rotations were determined by trial and error using the parameter FPEAK.

In the DFPSS data set, only fine particles were collected and measured. The six-factor results after rotation are presented. Accompanying the factors, individual error estimates were also computed for all of the factor elements. Figure 1a shows the resolved source profiles (the logarithmic scale profiles are shown in Figure 1b), and the associated temporal variations of the six possible sources are shown in Figure 2.

Sources 1 and 2 represent the C-rich/Si-Ca-Fe and C-rich/K aerosol sources, respectively. Both of these two sources produced high concentrations of carbonaceous particles through local combustion. By examining the temporal variations of these two sources, it was found that both sources had higher concentration peaks during the winter time, although the C/K source had secondary peaks during the summer time. Source 1 has dominant high concentrations of organic carbon (OC) and elemental carbon (EC) tailed by some of the soil dust components, Si, Ca and Fe. It is most probably the motor vehicle

emission source [15]. On the other hand, the biomass burning was likely to have contributed to the C/K source. The strong winter concentration peaks of these two sources are generally in agreement with other studies about Phoenix aerosol [16,17]. Carbonaceous particles are the primary cause of the urban haze that forms almost daily in Phoenix during the winter months when late night/early morning temperature inversions concentrate the natural and anthropogenic pollutants in the surface mixing layer. This results in a white to brown haze which can be easily seen in the shallow boundary layer during the early morning hours. It appears that sources 1 and 2 are closely associated with this winter haze phenomenon.

Source 3 is characterized by the presence of S, Cu, Zn, and Pb. It was identified as copper smelters. The temporal variation of this factor features high peaks in the winter time.

Source 4 is dominated by Na and Cl, a sea salt aerosol source. This factor does not exhibit a particular seasonal variation pattern.

Source 5 is represented by Al, Si, Ca, Ti and Fe, a typical soil source. The ubiquitous unpaved roads, construction sites, and tracts of farmland interspersed among the urban development would tend to produce particles of widely varied compositions of these typical crustal elements. They must have contributed to this factor.

Source 6 is a S-factor, characterized by the high peak of S. It is most probably attributed to the secondary sulfate particles emitted from a coal power plant. The seasonal variation of this factor features high concentration peaks during the summer time periods. Other studies also observed the similar trend for the sulfate particles [16,17]. For Phoenix aerosol, S is not an important component during the winter haze periods. The haze episodes are usually associated with carbon- and nitrate-rich aerosols, while in summer time the concentration of sulfate-rich aerosol is higher.

The investigation of different numbers of factors employed in the PMF analysis indicated that the 5-factor solution did not produce the sea salt factor and that the 7 or more-factor solutions gave 2 or more C-rich/Si-Ca-Fe factors which could be combined into one. Thus, the above 6-factor results have been presented.

To quantitatively estimate the mass contributions of the 6 resolved sources, the fine PM (particulate matter) mass was regressed against the factor scores using multiple linear regression (MLR). The constant of the linear regression was assumed to be zero. This regression process also provided an additional test for the PMF model and the appropriate number of factors that had been chosen for the analysis. An unrealistic number of factors for the PMF model very often resulted in negative values for the multiple linear regression coefficients. For the 6-factor solution, the obtained regression coefficients were all positive values with very small standard errors. The reconstructed mass concentrations of the

981 samples from the factor scores and the regression coefficients versus the observed fine PM mass concentrations are shown in Figure 3. The correlation coefficient is 0.96, indicating statistically the observed PM mass concentrations were represented quite well by the resolved 6 factors.

From the factor scores and the regression coefficients, the mass concentrations of each factor contributing to the total fine mass were obtained. Figure 4 presents the arithmetic means of the total fine mass concentrations for all of the 6 factors, and the seasonal means (summer: April-September; winter: October-March) are also shown. It can be seen that on average the motor vehicle and biomass burning factors contributed to the total fine PM mass concentrations the most, and that the sea salt and copper smelter factors contributed the least. Season-wise, the motor vehicle, biomass burning, and copper smelter factors contributed to the fine PM mass more in the winter time than in the summer time. The sea salt and soil factors did not show much seasonal variation, while the S-factor had higher mass contributions in the summer time.

### **Dichot Data**

The Dichot data set consisted of two parts, the fine and coarse fractions. The first effort was made to compare the PMF results on the Dichot fine fraction data with those on the DFPSS data. Here the 217x44 data matrix was used as the basis to infer the source profiles since the Na and Mg concentrations were not measured. Figure 5 shows sources profiles for the resolved six possible sources, and the associated source contributions are shown in Figure 6. It can be seen that the six source profiles and their temporal variations are quite similar to those obtained based on the DFPSS data. The multiple linear regression was again conducted to estimate the source contributions. The reconstructed mass concentrations of the 217 samples from the factor scores and the regression coefficients versus the observed fine mass concentrations are shown in Figure 7. The overall and seasonal (summer, winter) average contributions are shown in Figure 8. The overall and seasonal average contributions for the Dichot and DFPSS samples match quite well except for Soil and Sea Salt. Soil and Sea Salt contributed higher concentrations in the Dichot samples than in DFPSS samples.

To examine the possible reason for the high concentration of these two sources, Al and Si which are the major contributor for Soil source, and Cl which is the major contributor for Sea Salt source were considered. Figure 9 is a plot of Si, Al and Cl concentrations in Dichot and DFPSS samples that were found in the original data for the same days. This figure showed that there are two trend lines for Al and Si concentration. It was found that there is a time dependence on these trend lines. The concentration of Si and Al were higher before 20 Dec 1996 in the DFPSS sample which was due to installation of new



inlet. The PM<sub>2.5</sub> cyclone on the DFPSS was replaced with a WINS impactor. The shape of cut-point is sharper for the WINS compared to the cyclone inlet. This new inlet improved the separation of fine particles from the coarse particles by reducing the amount of soil in the fine particles samples. The overall average contributions for the sources were plotted in Figure 10 showing the concentration of soil before and after the installation of the new inlet. The figure showed that the overall average contribution of soil matched quite well after the installation of the new inlet. Figure 9 also showed that there is a factor of 1 to about 2 in Cl concentration between DFPSS and Dichot samples. This difference in the two concentration may contributed to the high concentration in the overall contributions of the sea salt in Dichot sample. In addition to the high concentration of OC and EC in the Sea Salt source profiles in the Dichot source profiles in Figure 5 compared to the Sea Salt source profile in the DFPSS in Figure 1.

For the coarse fraction data, the five-factor model will be discussed as it gave the most satisfactory results in terms of explaining the underlying structure in the data. Figure 11 shows the sources profiles of the five possible sources, and the associated source contributions are shown in Figure 12. The first source is a typical sea salt source due to the high peak of Cl. The second factor is the soil dust source due to Si and crustal elements such as Al, Ti, and Fe. Si-rich particles have previously been reported mainly in the coarse particles (15). In Phoenix, airborne soil particles derived primarily from igneous and metamorphic rocks are transported into the metropolitan area by strong winds from southern and eastern Arizona and northern Mexico (17). Major sources for soil derived particles are paved and unpaved roads, agriculture, and construction (2,18). The third factor is Fe source/motor vehicle due to Fe, S, and Pb. The fourth factor is a Ca rich source which can be produced by cement manufacture, by traffic on concrete roads or by some construction activities near the sampling site. The presence of S in Ca-rich source is suggestive of gypsum ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) or anhydrite ( $\text{CaSO}_4$ ). Gypsum and anhydrite particles can originate from weathering of natural material. The fifth factor is characterized by S, La and other trace metal elements which may be due to a coal power plant or other industrial source.

The coarse fraction mass concentration was regressed against the factor scores using multiple linear regression to estimate the source contributions. The reconstructed mass concentrations of the 217 samples from the factor scores and the regression coefficients versus the observed coarse mass concentrations are shown in Figure 13. The r-squared is 0.98 indicating that the 5 factors represent the measured coarse mass concentrations very well. It was found that the major contributing sources were the soil dust and the construction (Ca-rich) sources as shown in Figure 14. This match quite well with previous Phoenix aerosol studies which showed that the coarse fraction of the Phoenix aerosol is dominated by mineral particles, mostly Si-Rich, as shown by both bulk and individual particles analysis

methods (15,17, 18). These sources contributed much of the coarse aerosol mass through emitting Al, Si, Fe, and Ca particles, having higher concentrations in winter time.

## **Conclusions**

Aerosol chemical composition data for the fine and coarse particles from Phoenix, AZ have been analyzed using the positive matrix factorization (PMF) method. It is concluded that PMF was an effective method of identifying possible aerosol sources. Through multiple linear regression using the measured particulate matter (PM) mass concentrations, the mass contributions of the resolved sources have been estimated. Six and five source factors have been obtained for the fine and coarse particles, respectively. Among them, the motor vehicle and biomass burning factors, two major sources producing carbonaceous particles, contributed to the fine PM mass the most, and contributed more in the winter time than in the summer time. The other sources for the fine particles are copper smelter, sea salt, soil, and S-factor. While for the coarse particles, the major contributing sources were the soil dust and the construction (high Ca) sources. The other sources were the sea salt, Fe source/motor vehicle and coal power plant sources. The results of this study are significant in helping better understanding of the origins and mass contributions of aerosol sources in Phoenix.

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## Figure Captions

- Figure 1a. Source profiles (linear scale) with  $Q = 50878$ ,  $FPEAK=0.20$  for DFPSS Sampler
- Figure 1b. Source profiles (log scale) with  $Q = 50878$ ,  $FPEAK=0.20$  for DFPSS Sampler.
- Figure 2. Time series of estimated source contributions to the DFPSS Sample masses.
- Figure 3. Predicted fine particle mass concentrations compared with the measured concentration values for the DFPSS data.
- Figure 4. Average mass contributions to the DFPSS measured fine particle masses for winter, summer and annual time periods.
- Figure 5. Source profiles for the DICHOT fine particle data with  $Q = 8875$ ,  $FPEAK=0.15$
- Figure 6. Source contributions for the DICHOT fine particle data with  $Q = 8875$ ,  $FPEAK=0.15$ .
- Figure 7. Predicted fine particle mass concentrations compared with the measured concentration values for the DICHOT data.
- Figure 8. Average mass contributions to the DICHOT measured fine particle masses for winter, summer and annual time periods.
- Figure 9. Comparison of DFPSS and DICHOT concentration data for aluminum, silicon, and chlorine.
- Figure10. Comparison of the average source contributions for the DFPSS and DICHOT fine particle data.
- Figure 11. Source Profiles for the DICHOT coarse particle samples
- Figure 12. Time series of source contributions for the DICHOT coarse particle samples.
- Figure 13. Average mass concentration of each Source DICHOT Coarse Samples
- Figure 14. Observed vs Predicted Mass Concentrations for the DICHOT Coarse Samples

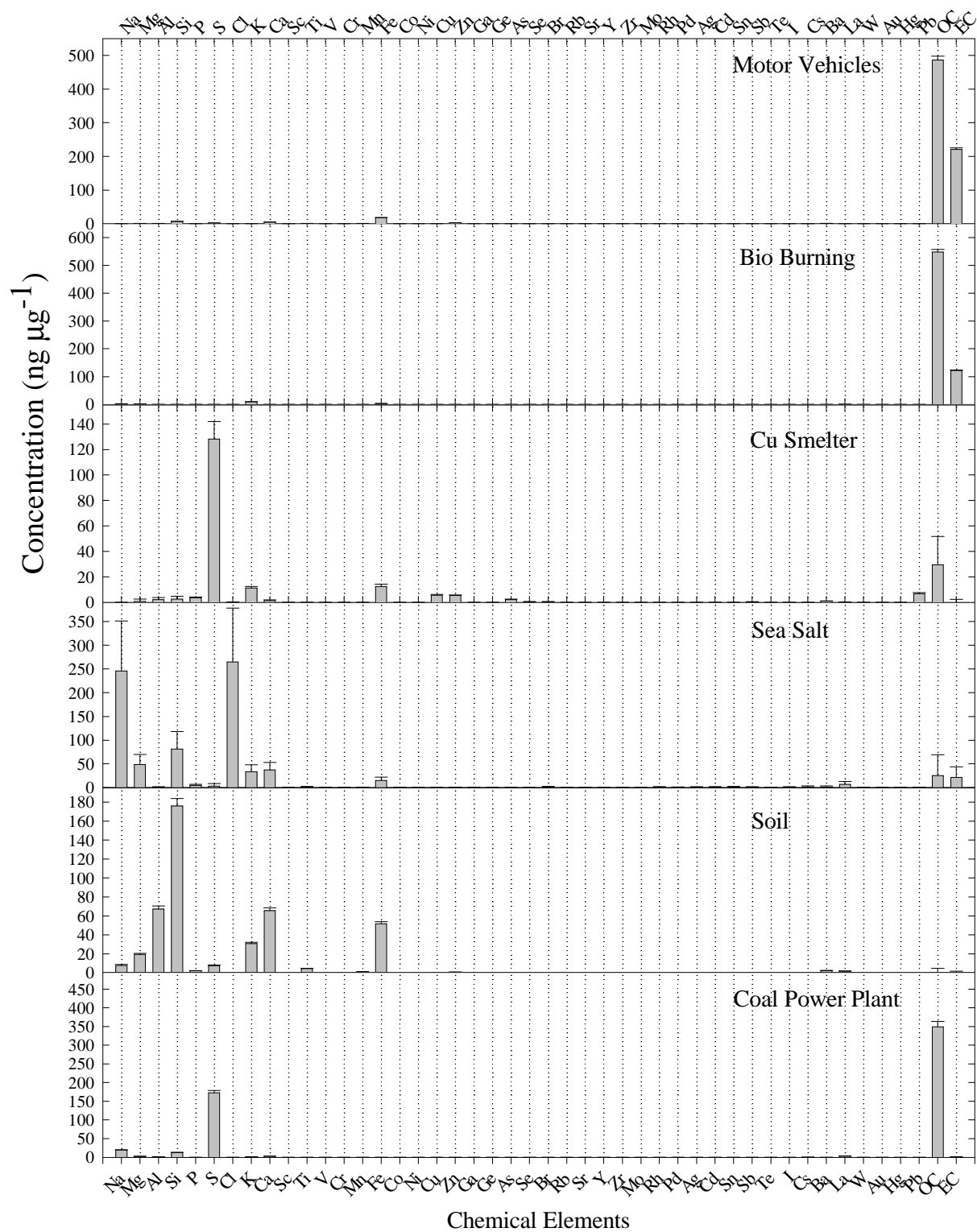


Figure 1.

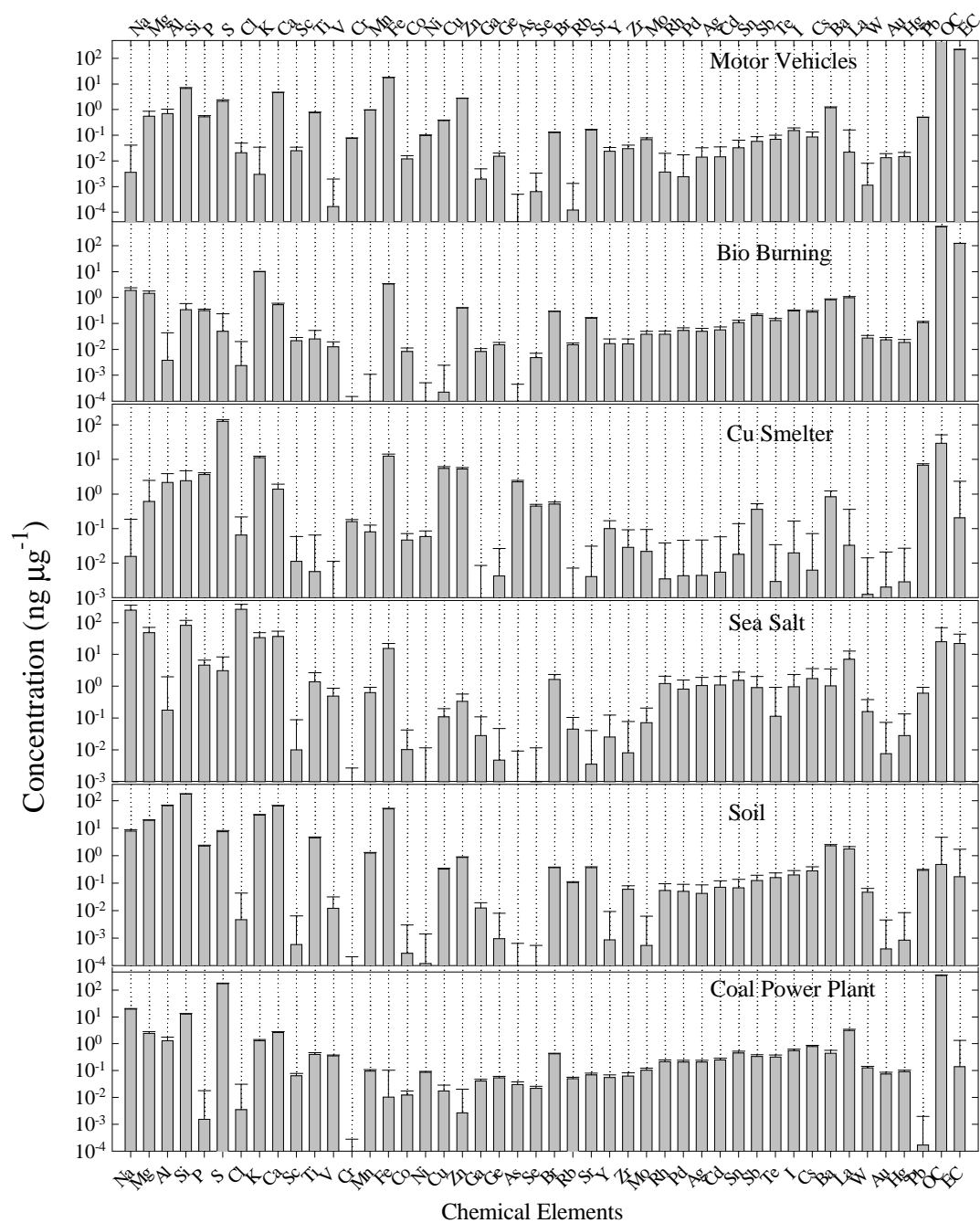


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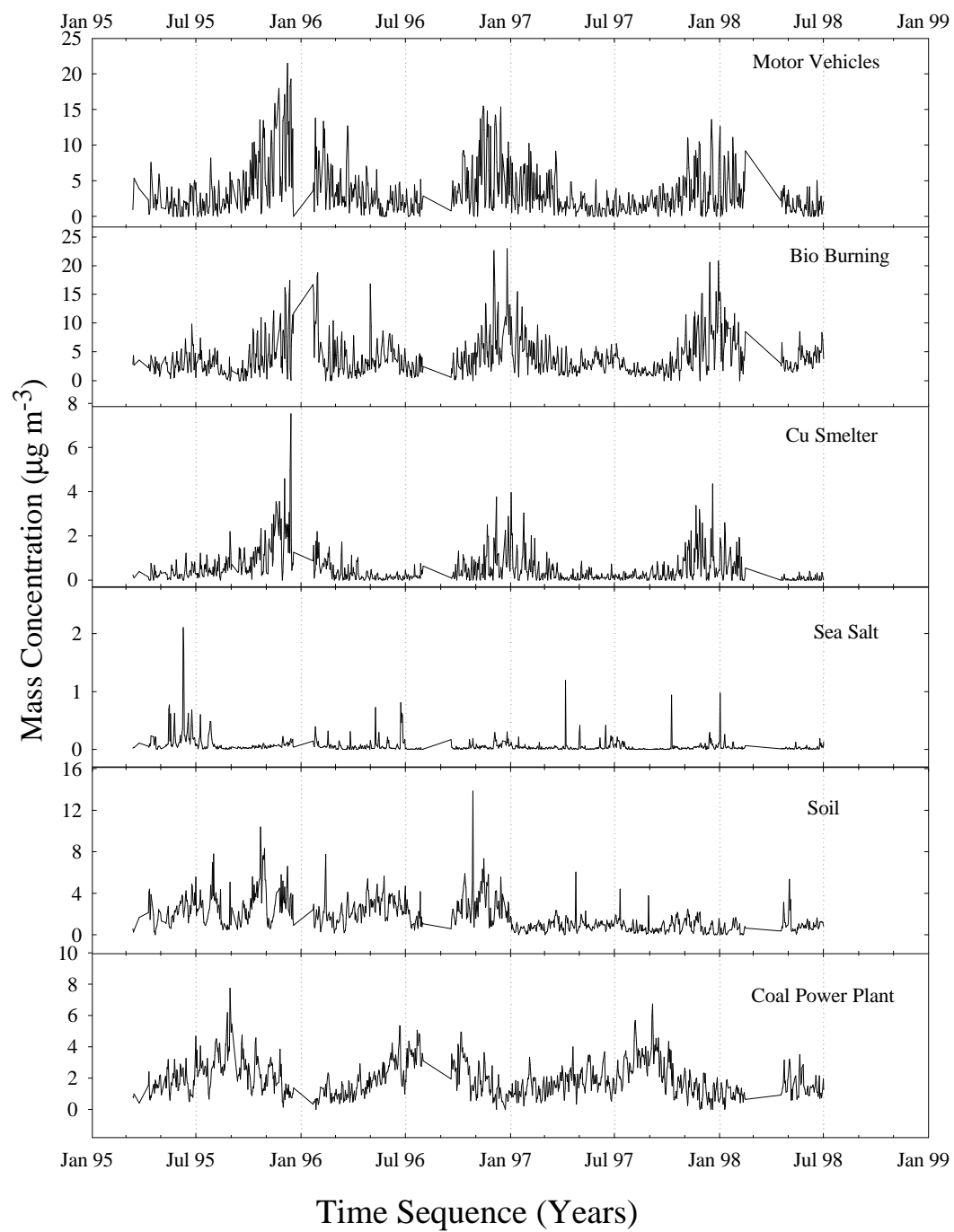


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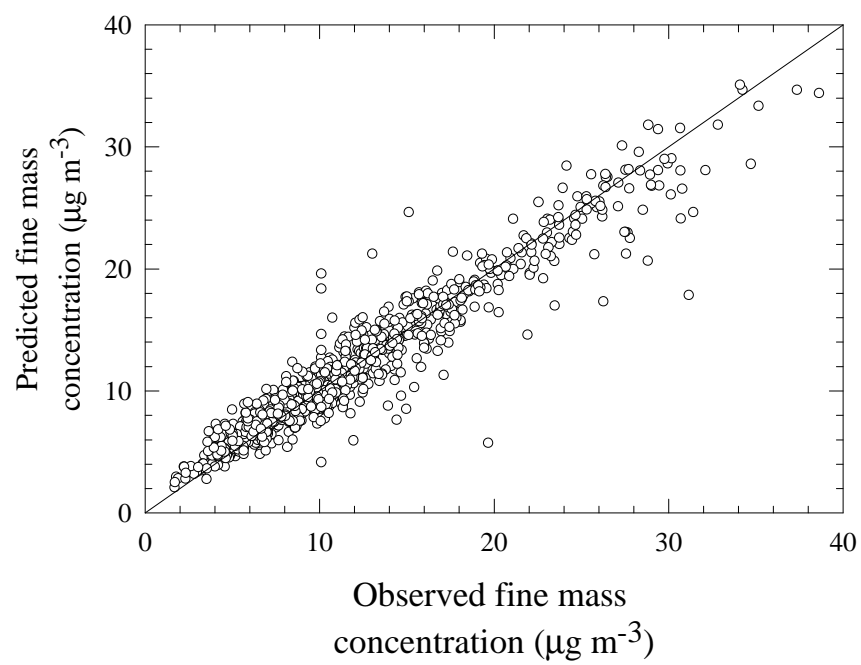


Figure 3.



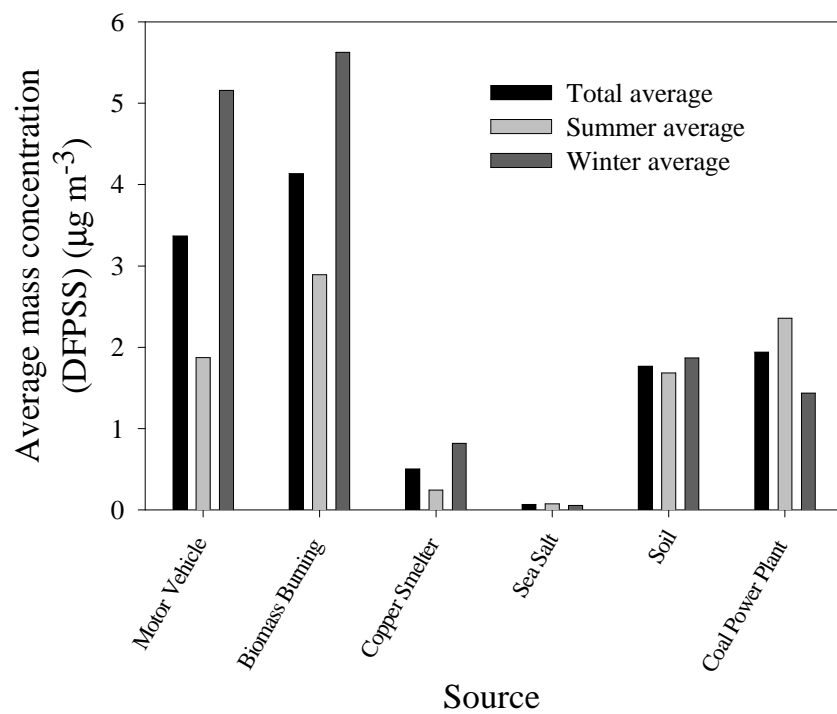


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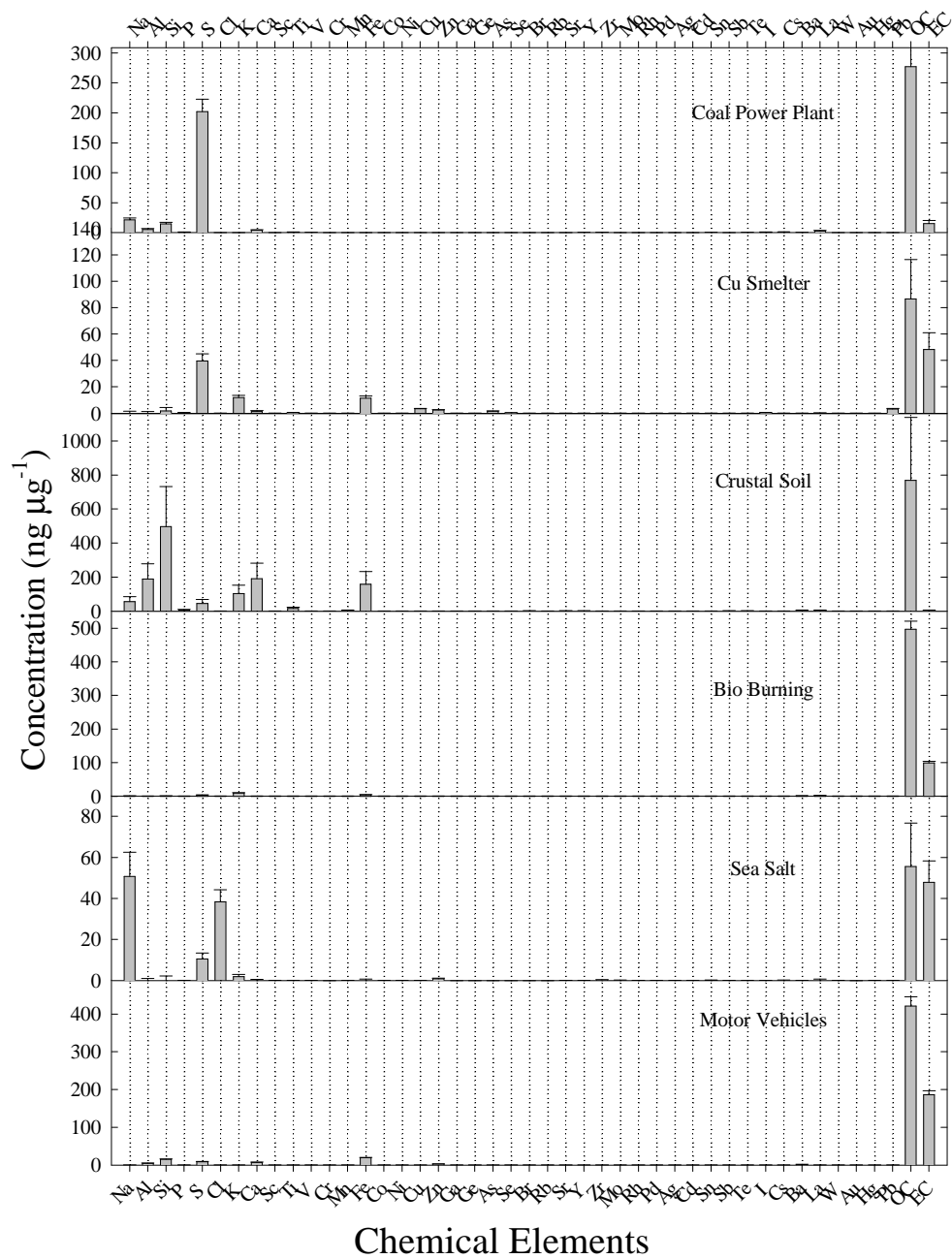


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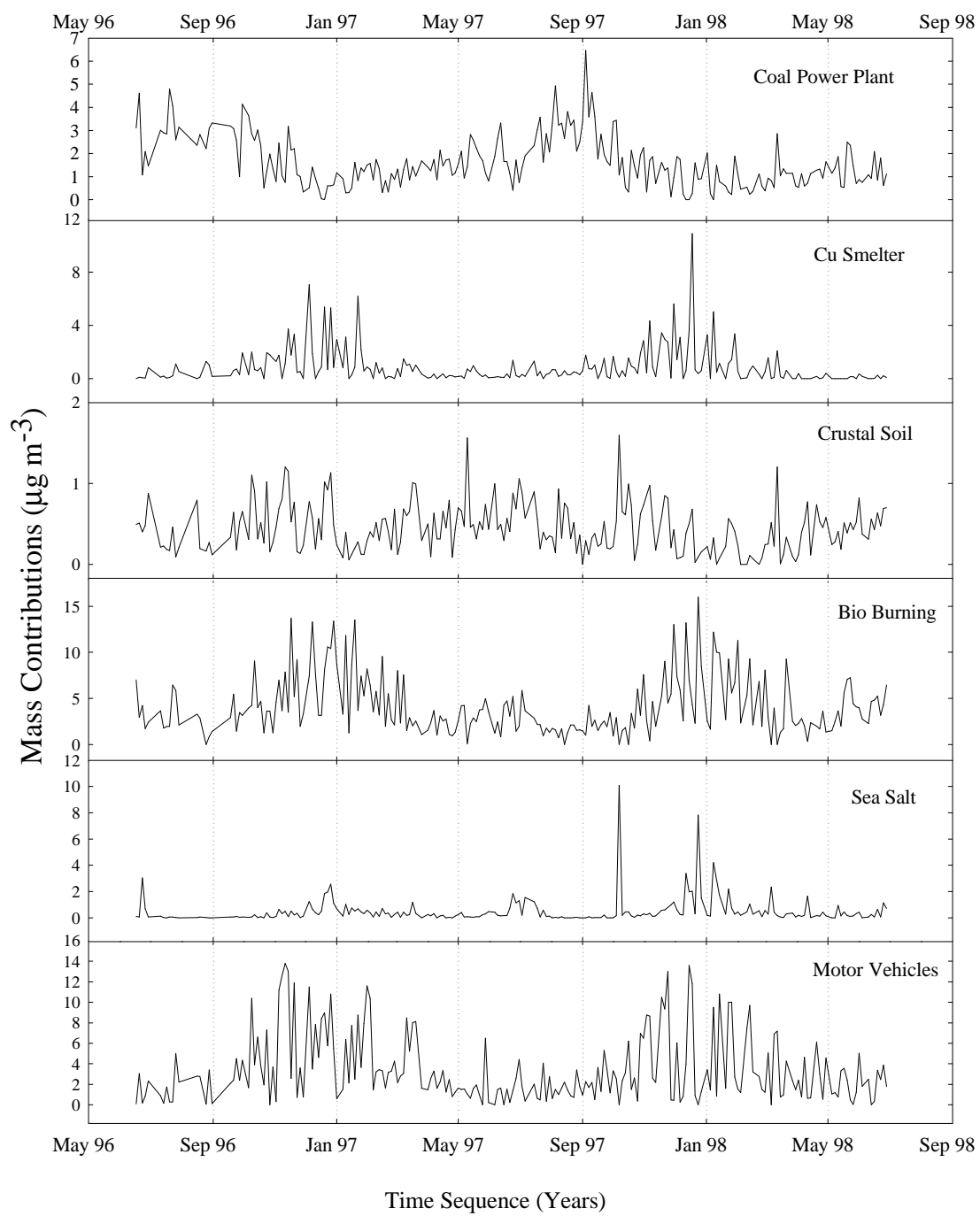


Figure 6.

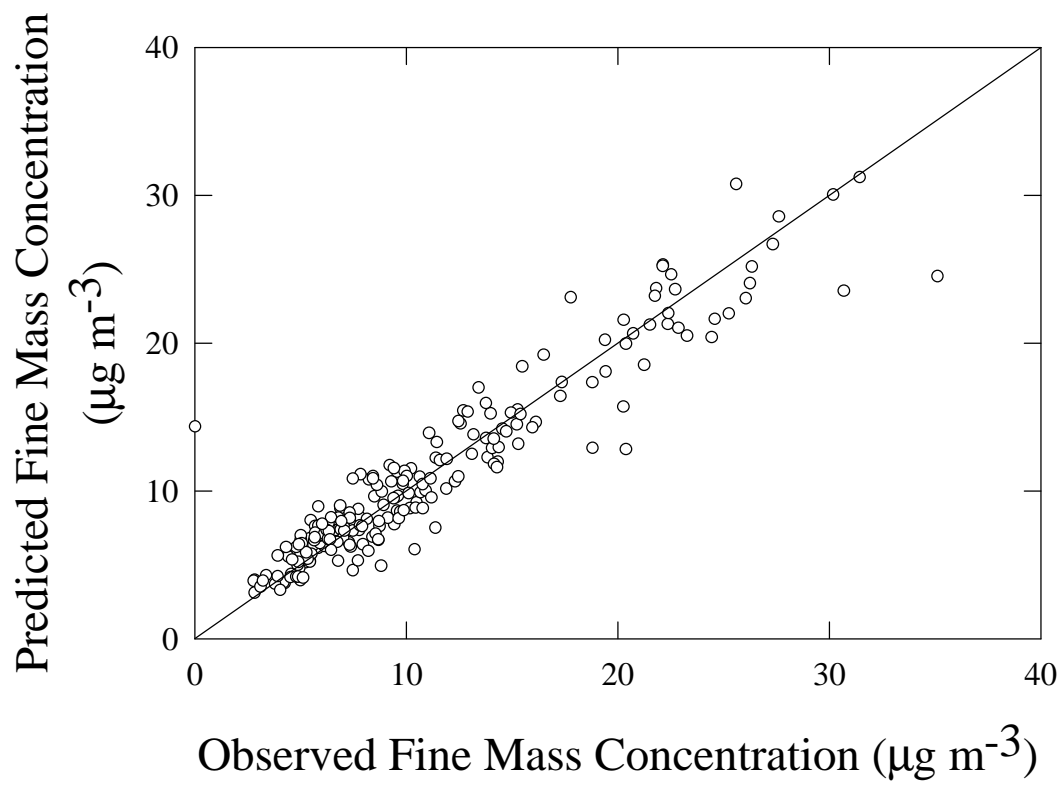


Figure 7.

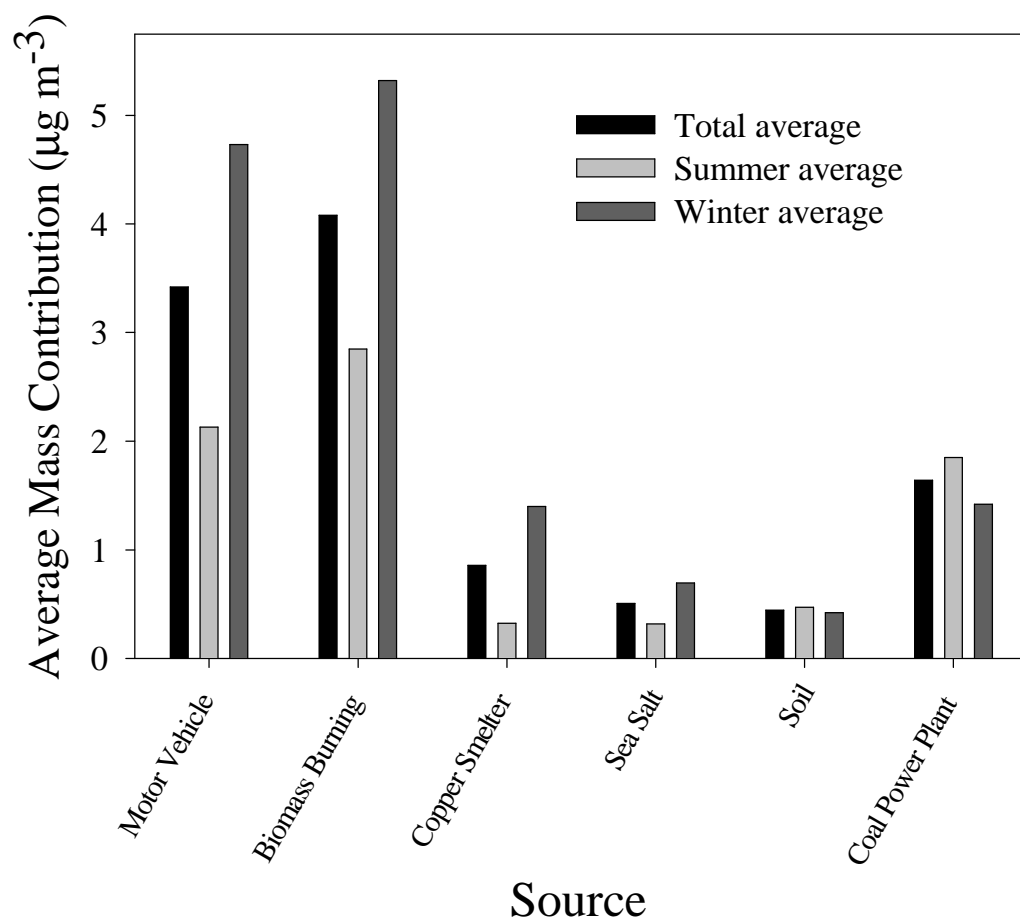


Figure 8.

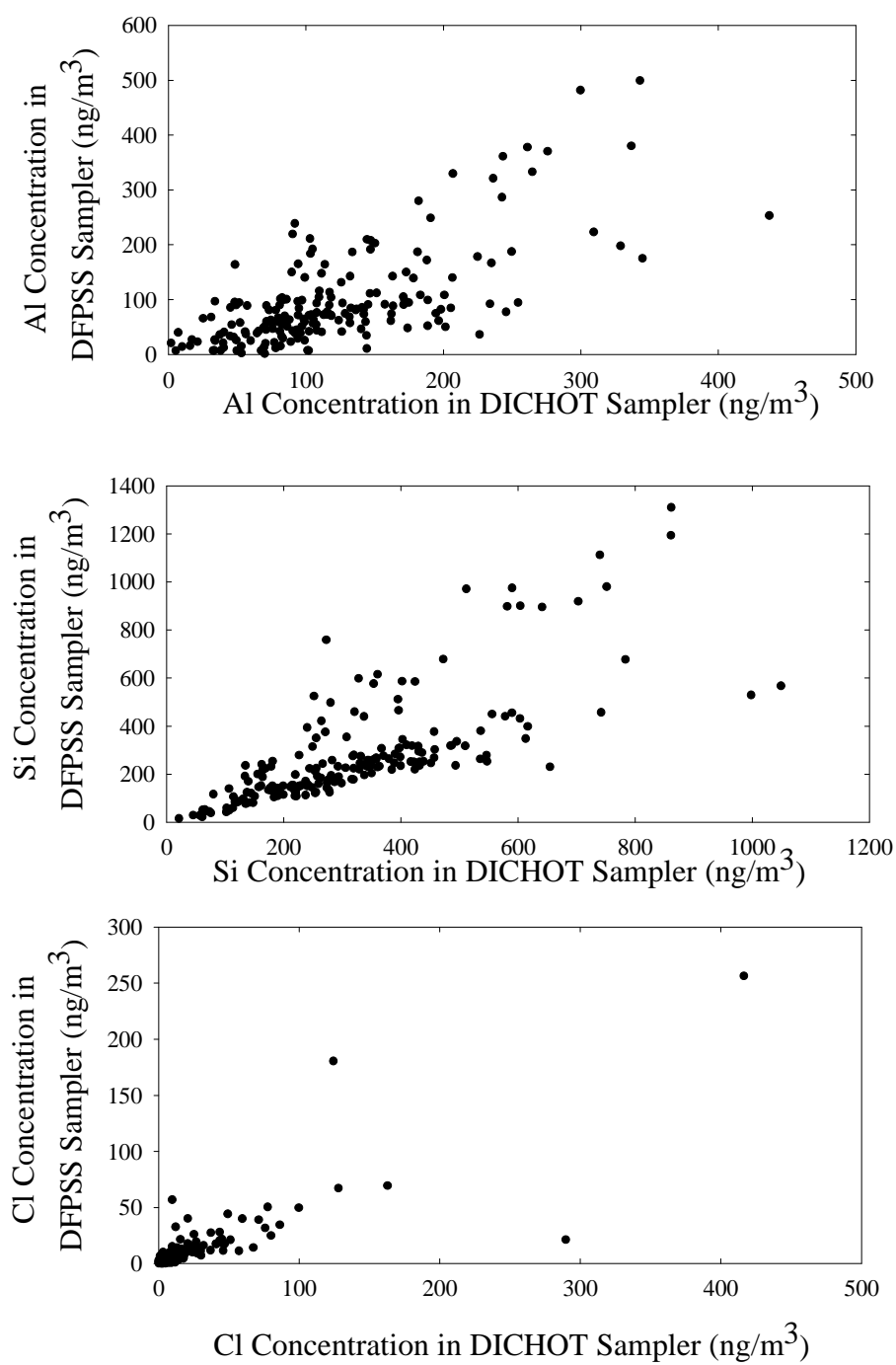


Figure 9.

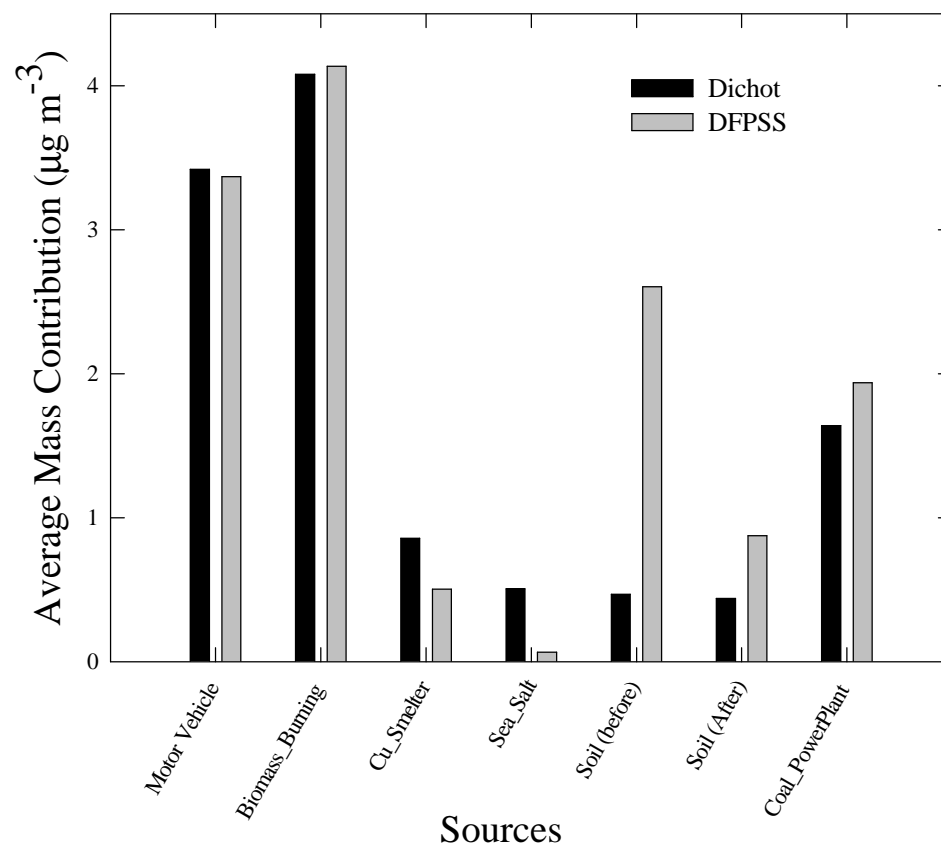


Figure 10.

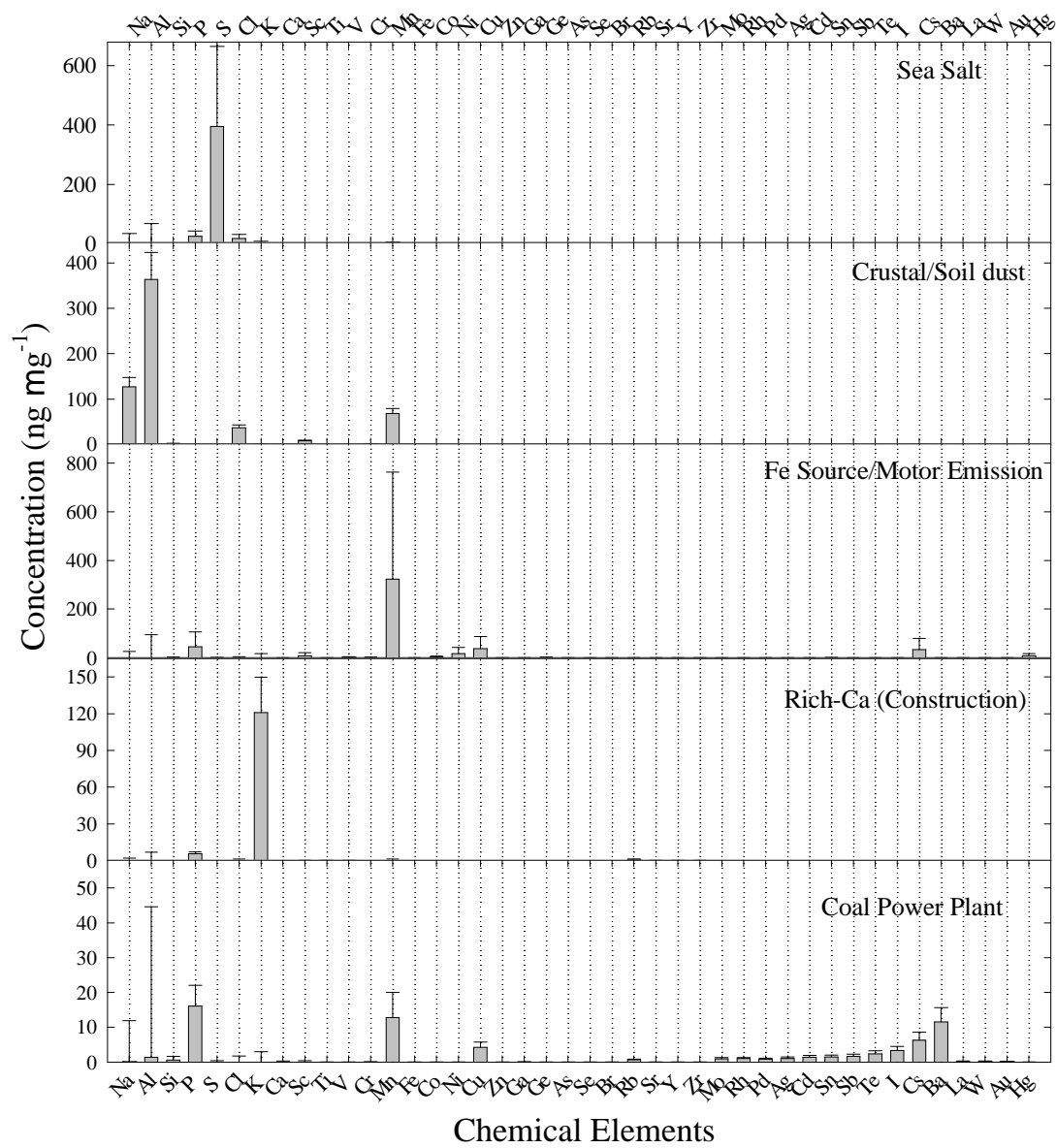


Figure 11.



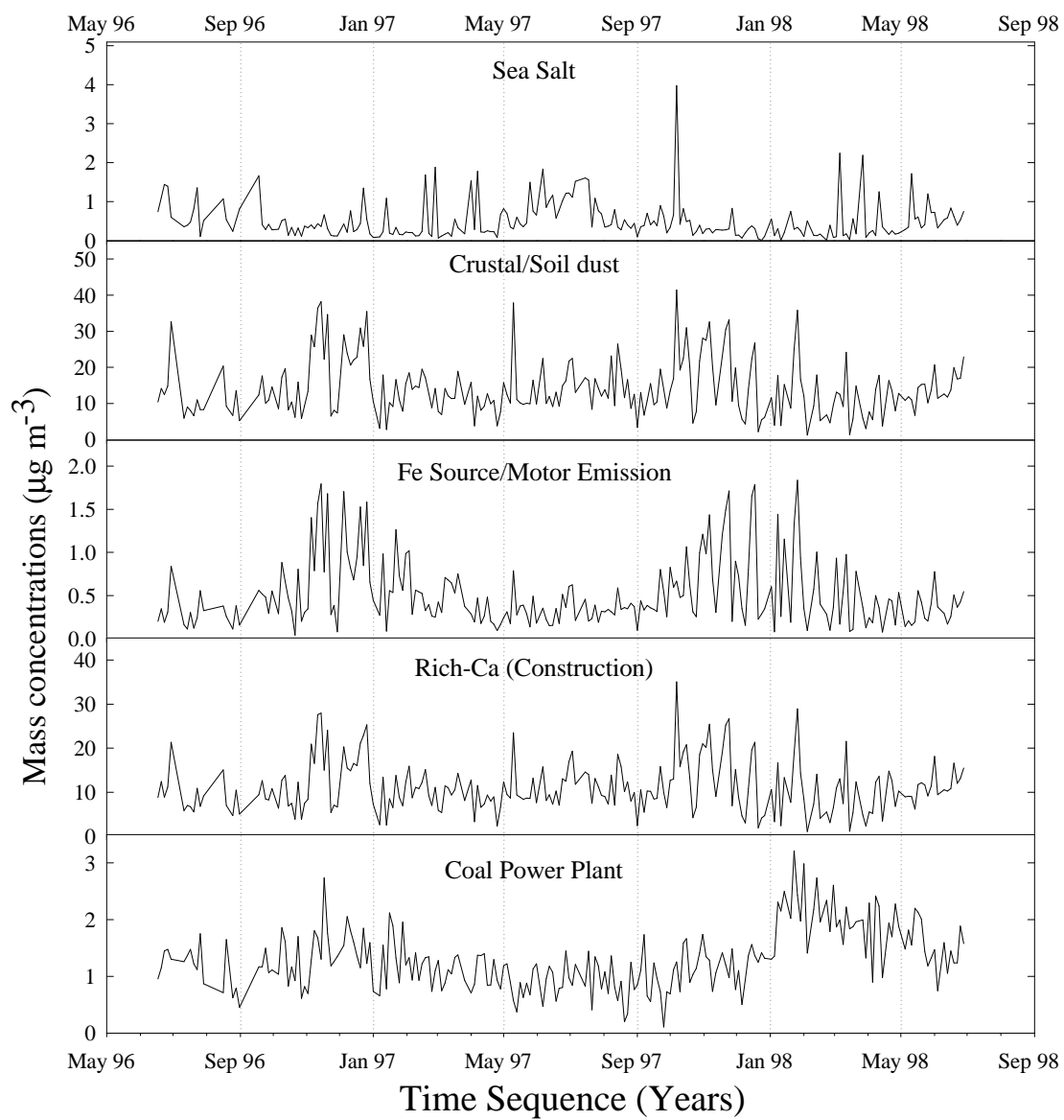


Figure 12.

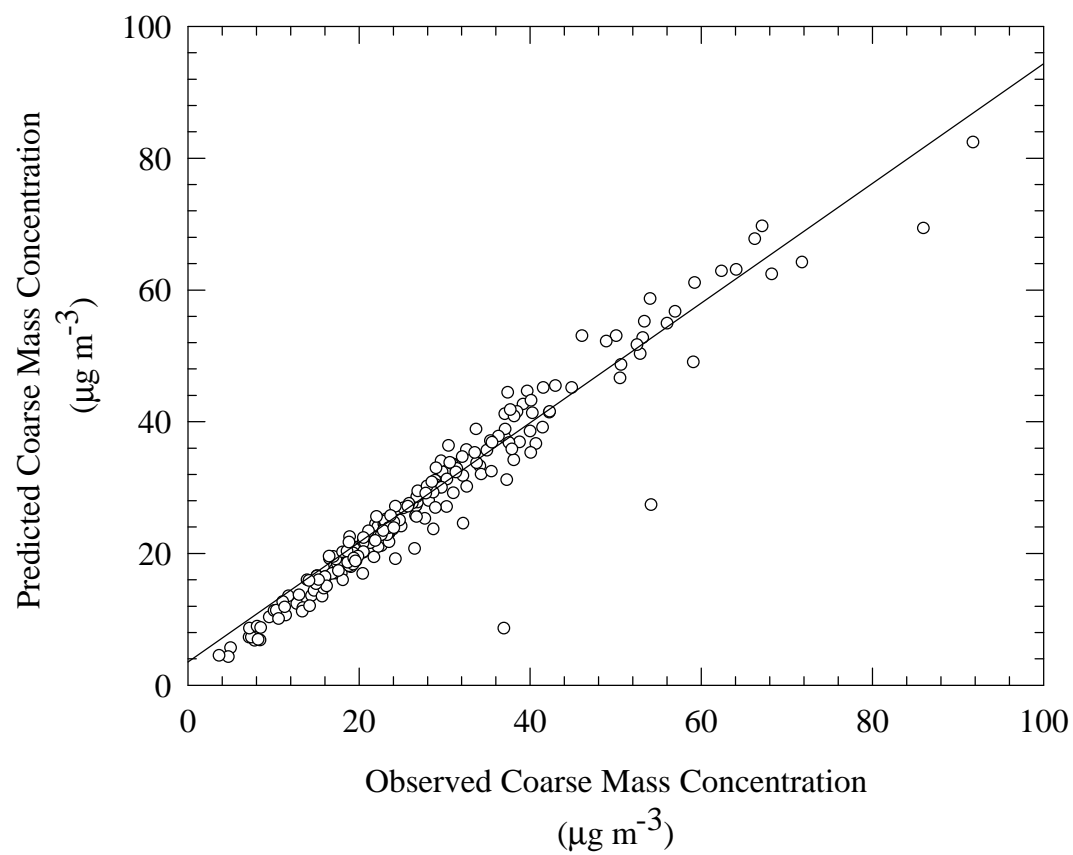


Figure 13.

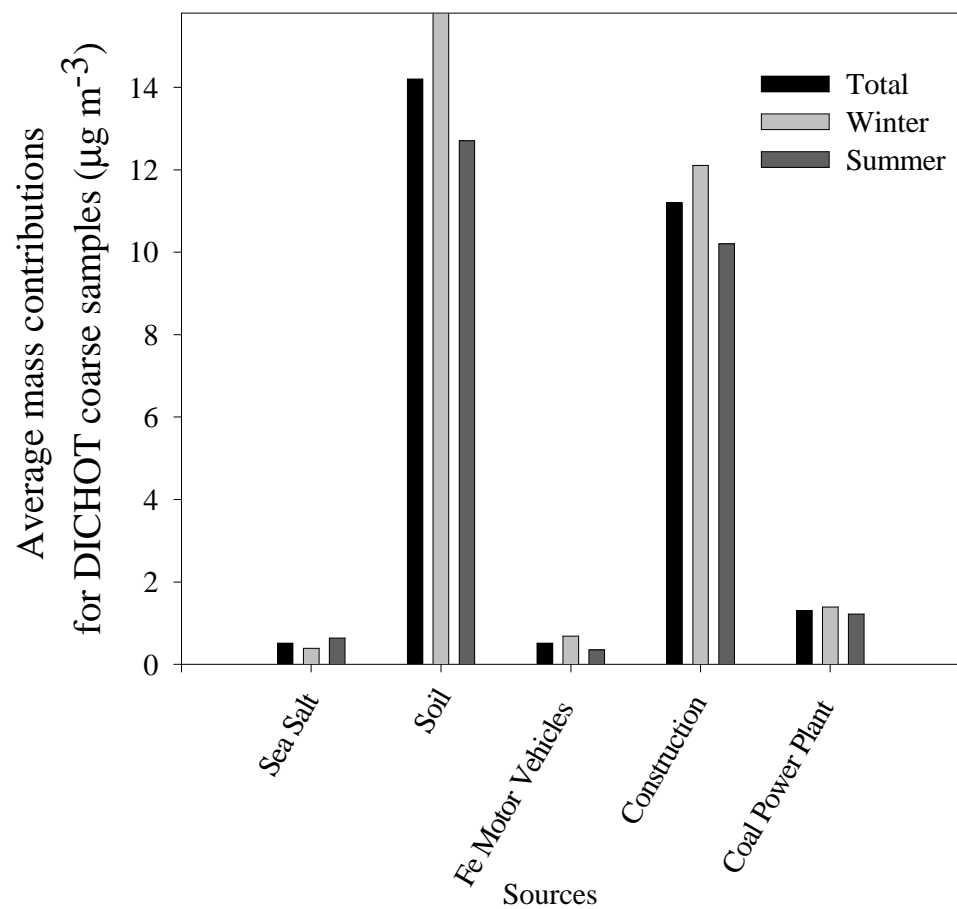


Figure 14.